

All-order calculations of the spectra of superheavy elements E113 and E114

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We apply a recently developed method (V. A. Dzuba, PRA **90**, 012517 (2014); J. S. M. Ginges and V. A. Dzuba, PRA **91**, 042505 (2015)) to calculate energy levels of superheavy elements Uut ($Z = 113$), Fl ($Z = 114$), and Fl⁺. The method combines the linearized single-double coupled-cluster technique, the all-order correlation potential method and configuration interaction method. Breit and quantum electrodynamic corrections are included. The role of relativistic and correlation effects is discussed. Similar calculations for Tl, Pb and Pb⁺ are used to gauge the accuracy of the calculations.

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I. INTRODUCTION

The study of the superheavy elements is an important area of research motivated by the predicted “island of stability” in the region $Z > 104$. Elements with nuclear charge up to $Z = 118$, have been synthesised (see, e.g., Refs. [1–5]), and evidence for naturally-occurring E122 was reported [6].

Apart from huge activity in the theoretical and experimental nuclear physics there are also many theoretical works in atomic physics and quantum chemistry with attempts to predict the chemical properties of the superheavy elements and their electron structure and spectra (see, e.g. [7–9]).

Superheavy elements E113 and E114 are of special interest due to their closeness to the hypothetical island of stability and relatively simple electron structure. The E113 atom can be considered as a system with one external electron above closed-shell core which ends with the $7s^2$ subshell. Its lighter analog is Tl. The E114 atom can be considered as a system with two valence electrons. There is a number of calculations of electron spectra of elements E113 and E114 using multi-configuration Dirac-Fock, coupled-cluster, configuration interaction methods and their combinations [10–17]. The results of different approaches agree in general trends caused by interplay of relativistic and correlation effects. However, actual numbers for the energies often differ beyond the uncertainty claimed by the authors. Therefore, it is important to redo the calculations using the most advanced techniques which should lead to more accurate and reliable results. In present paper we apply the recently developed technique [18] which combines the all-order correlation potential method [19], supplemented by ladder diagrams [20] with the configuration interaction method [21]. The technique gives very accurate results for energy levels of Cs, Tl, Ba, Lu, Ra and those ions of these elements which have one or two valence electrons above closed shells [18, 20]. It was used to calculate energy levels of superheavy elements E119, E120 and E120⁺ [22–24]. We demonstrate that the method also works for Pb and

Pb⁺. Then we apply it to calculate energy levels of E113, E144 and E144⁺.

II. METHOD OF CALCULATIONS

The method was described in detailed in our previous papers [18, 20, 23, 24]. Here we repeat its main points with the focus on the details specific for current calculations.

A. Atoms with one valence electron

Calculations are done in the V^{N-1} approximation which means that the self-consistent potential is formed by the $N - 1$ electrons of the closed-shell core (the V^{N-1} potential). A complete set of the single-electron orbitals is obtained by solving the equations

$$h_0\psi_0 = \epsilon_0\psi_0, \quad (1)$$

using the B-spline technique [25, 26]. Here h_0 is the relativistic Hartree-Fock Hamiltonian

$$h_0 = c\boldsymbol{\alpha} \cdot \mathbf{p} + (\beta - 1)mc^2 - \frac{Ze^2}{r} + V^{N-1}. \quad (2)$$

B-spline basis set and Feynman diagram technique are used to calculate the all-order correlation potential (CP) $\hat{\Sigma}$ [19, 20]. The CP operator $\hat{\Sigma}$ is defined in such a way that its expectation value for a valence state v is equal to the correlation correction to the energy of this state: $\delta\epsilon_v = \langle v|\hat{\Sigma}|v\rangle$. Perturbation theory expansion for $\hat{\Sigma}$ starts from the second order, we use the $\hat{\Sigma}^{(2)}$ notation for corresponding CP. Then we include three classes of the higher-order correlations into the all-order CP $\hat{\Sigma}^{(\infty)}$ [19]: a) screening of Coulomb interaction, b) hole-particle interaction, and c) ladder diagrams [20]. States and energies of the valence electron are found by solving the equation [27]

$$(\hat{h}_0 + \hat{\Sigma})\psi_v = \epsilon_v\psi_v. \quad (3)$$

TABLE I: Energy levels of Tl and Pb^+ calculated in different approximations. Final results are the sum of Σ^∞ and ladder contributions. Δ is the difference between final theoretical results and experimental numbers. The results for Tl are taken from Ref. [20]. The results for Pb^+ obtained in present work. Experimental numbers are taken from the NIST database [28].

State	RHF	$\Sigma^{(2)}$	Σ^∞	Ladder	Final	Δ	Expt.
Tl							
$6p_{1/2}$	42823	51597	50815	-1215	49600	336	49264
$6p_{3/2}$	36636	43524	42491	-794	41697	226	41471
$7s_{1/2}$	21109	23375	22887	-43	22844	58	22786
Pb^+							
$6p_{1/2}$	114360	123612	122547	-1421	121126	-119	121245
$6p_{3/2}$	100731	109451	108108	-987	107121	-43	107164
$7s_{1/2}$	58660	62793	61895	-104	61791	-5	61796

Here $\hat{\Sigma}$ can be either the second-order CP $\hat{\Sigma}^{(2)}$ or all-order CP $\hat{\Sigma}^{(\infty)}$. Note that by iterating the Eq. (3) we include one more class of higher-order correlations, the iterations of $\hat{\Sigma}$ (contributions proportional to $\hat{\Sigma}^2$, $\hat{\Sigma}^3$, etc.). The wave-functions ψ_v of the valence electron found by solving the Eq. (3) are often called Brueckner orbitals. Corresponding energies ϵ_v include correlations. Breit and quantum electrodynamic corrections are also included (see below).

Table I presents the results of the calculations for the low s and p states of Tl and Pb^+ . The results for Tl are taken from our earlier paper [20], the results for Pb^+ are obtained in this work. Contributions of the ladder diagrams are presented separately because it is the latest addition to the method and it is important to emphasize its role. Tl and Pb^+ have similar electron structure, therefore it is natural to expect that the results are also similar. We see however that the results for Pb^+ are even slightly better than for Tl. This is probably due to stronger Coulomb potential leading to smaller relative value of the correlation correction. Indeed, the correlation correction to the energy is equal to the difference between the relativistic Hartree-Fock results (the RHF column in Table I) and the experimental values. We see that the absolute value of the correlation correction is larger for Pb^+ while the relative value is smaller for Pb^+ than for Tl. In the end the accuracy for the energy is on the level of 0.5%.

B. Atoms with two valence electrons

We use the configuration interaction (CI) technique combined with the all-order methods to include core-valence correlations [18, 21, 23]. The effective CI Hamiltonian for the system of two valence electrons has the form

$$\hat{H}^{\text{CI}} = \hat{h}_1(r_1) + \hat{h}_1(r_2) + \hat{h}_2(r_1, r_2), \quad (4)$$

where \hat{h}_1 is the single-electron operator and \hat{h}_2 is the two-electron operator. The \hat{h}_1 operator is the sum of the RHF operator and the CP $\hat{\Sigma}_1$

$$\hat{h}_1 = \hat{h}_0 + \hat{\Sigma}_1. \quad (5)$$

Here the CP $\hat{\Sigma}_1$ is the all-order CP considered in previous section. We introduce index 1 to stress that this is a single-electron operator.

The \hat{h}_2 operator is the sum of Coulomb interaction and the correlation operator $\hat{\Sigma}_2$ [18]

$$\hat{h}_2(r_1, r_2) = \frac{e^2}{|r_1 - r_2|} + \hat{\Sigma}_2(r_1, r_2). \quad (6)$$

The $\hat{\Sigma}_2$ operator appear due to core-valence correlations and can be understood as screening of Coulomb interaction between valence electrons by core electrons. This is also the all-order operator which comes from solving the single-double (SD) coupled-cluster equations [18]. Note that solving the SD equation produce both, single-electron CP $\hat{\Sigma}_1$ and two-electron correlation operator $\hat{\Sigma}_2$. However, for many atomic systems, including those considered in present work and those considered previously [18, 24], using the all-order CP $\hat{\Sigma}^{(\infty)}$ which was discussed in previous section, leads to better results than using the SD operator $\hat{\Sigma}_1$.

Table II shows the results of the calculations for Pb. We present energies and g -factors and compare them to the experiment. The g -factors are useful for identification of the states. Comparison with experiment shows that the accuracy for the energies is on the level of 1-2% or better.

C. Breit and QED correction

Since we are considering heavy atoms it is important to include Breit and quantum electrodynamic (QED) corrections.

The Breit operator in the zero energy transfer approximation has the form:

$$h^B = -\frac{\boldsymbol{\alpha}_1 \cdot \boldsymbol{\alpha}_2 + (\boldsymbol{\alpha}_1 \cdot \mathbf{n})(\boldsymbol{\alpha}_2 \cdot \mathbf{n})}{2r}, \quad (7)$$

where $\mathbf{r} = \mathbf{n}r$, r is the distance between electrons, and $\boldsymbol{\alpha}$ is the Dirac matrix.

We use the radiative potential method introduced in Ref. [29] to include QED corrections to the energies. The radiative potential has the form

$$V_{\text{rad}}(r) = V_U(r) + V_g(r) + V_e(r), \quad (8)$$

where V_U is the Uehling potential and V_g is the potential arising from the magnetic formfactor, and V_e is the potential arising from the electric formfactor.

Both, Breit and QED operators are included to the Hartree-Fock iterations so that an important relaxation effect is taken into account [30–32].

TABLE II: Calculated excitation energies (E , cm^{-1}), and g -factors for lowest states of Pb atom.

State	This work		Experiment	
	E	g	E	g
$6p^2$ 1S_0	0	0.0000	0	0.0
$6p^2$ 3P_1	7922	1.4999	7819	1.501
$6p^2$ 3D_2	10940	1.2916	10650	1.269
$6p^2$ 3D_2	21924	1.2085	21458	1.230
$6p^2$ 1S_0	29177	0.0000	29467	0.0
$7s6p$ $^1S_0^o$	35109	0.0000	34959	0.0
$7s6p$ $^3P_1^o$	35536	1.3509	35287	1.349
$6p7p$ 3D_1	43236	0.6707	42919	
$6p7p$ 1S_0	44449	0.0000	44401	0.0
$6p7p$ 3P_1	44873	1.4690	44675	
$6p6d$ $^3F_2^o$	44986	0.7962	45443	0.798
$6p7p$ 3D_2	44997	1.1739	44809	
$6p6d$ $^1P_1^o$	46132	0.8186	46068	0.864
$6p6d$ $^3D_2^o$	46162	1.2715	46061	1.247
$6p6d$ $^3F_3^o$	46324	1.1184	46328	1.116
$7s6p$ $^3P_2^o$	48765	1.4814	48188	1.496
$8s6p$ $^1S_0^o$	48784	0.0000	48726	0.0
$8s6p$ $^3P_1^o$	48811	1.3238	48687	1.304
$7s6p$ $^1P_1^o$	49892	1.1014	49440	1.131
$6p9p$ 3D_1	51422	0.6690	51321	
$6p9p$ 1S_0	51683	0.0000	51786	

TABLE III: Energy levels (in cm^{-1}) of superheavy elements Uut ($Z = 113$) and Fl $^+$ ($Z = 114$) calculated in different approximations. Notations like in Table I.

State	RHF	$\Sigma^{(2)}$	$\Sigma^{(\infty)}$	Ladder	Final	Other ^a
Uut						
$7p_{1/2}$	54901	61929	61953	-2183	59770	60154
$7p_{3/2}$	31557	38498	36623	-497	36126	34938
$8s_{1/2}$	22193	24653	23761	-32	23729	21313
Fl $^+$						
$7p_{1/2}$	130420	138110	138105	-2333	135772	137710
$7p_{3/2}$	89802	99170	96708	-667	96041	97329
$8s_{1/2}$	60844	65316	63832	-82	63750	63964

^aReference [17]

III. RESULTS AND DISCUSSION

Table III shows the results of calculations for Uut (E113) and Fl $^+$ superheavy elements in the same form as in Table I for Tl and Pb $^+$. Comparison shows some interesting trends. The total value of the correlation correction for superheavy elements and their lighter analogs are similar but slightly smaller for the superheavy elements. This is probably due to relativistic relaxation which leads to increased energy interval between core and valence states. On the other hand, the contribution of ladder diagrams is larger for the ground states of E113 and Fl $^+$ than for Tl and Pb $^+$. Ladder diagrams describe residual Coulomb interaction between valence electron and the core. Larger contribution probably reflects the fact that due to relativistic relaxation the su-

TABLE IV: Calculated excitation energies (E , cm^{-1}), and g -factors for lowest states of superheavy element Fl.

State	This work		Other	
	E	g	E^a	E^b
$7p^2$ 1S_0	0	0.0000	0	0
$7p^2$ 3P_1	26780	1.4995	27316	26342
$7p^2$ 3D_2	29462	1.1966	29149	28983
$8s7p$ $^1S_0^o$	43573	0.0000	44036	43111
$8s7p$ $^3P_1^o$	43876	1.3413	44362	43441
$7p8p$ 3D_1	51646	0.6670	51834	51302
$7p8p$ 1S_0	52724	0.0000	53149	52487
$7p8p$ 3P_1	54842	1.4932	55414	54647
$7p8p$ 3D_2	55015	1.1713	55191	54814
$7p7d$ $^3D_2^o$	55814	1.1780	56988	
$7p7d$ $^1D_2^o$	55828	0.8730	57413	
$7p7d$ $^3F_3^o$	55890	1.1138	57481	
$7p7d$ $^1P_1^o$	55910	0.8259	57244	
$9s7p$ $^1S_0^o$	57607	0.0000	57367	
$9s7p$ $^3P_1^o$	57663	1.3316		
$7p9p$ 3D_1	60198	0.6669		
$7p9p$ 1S_0	60324	0.0000		
$7p9p$ 3D_2	61272	1.1769	57413	
$7p8d$ $^3F_2^o$	61612	0.7717		
$7p6f$ 1D_2	61620	0.9097		
$7p6f$ 3G_3	61650	0.8357	60291	
$7p6f$ 3F_3	61653	1.1917	60298	
$7p6f$ 3G_4	61655	1.0838	60311	

^aReference [17]

^bReference [15]

perheavy elements in the ground state have smaller size than their lighter analogs. Since the total value of the correlation correction to the energies is very similar for heavy and lighter elements we expect that the accuracy of the calculations is also very similar, i.e. $\sim 0.5\%$.

The results of present calculations are in a reasonable agreement with previous SD+CI calculations of Ref. [17] (see Table III). However, they are closer to the results of coupled-cluster calculations of Ref. [10, 11]. This is true for both, ionization potential and excitation energies.

The results for Fl (E114) are presented in Table IV and compared with previous calculations of Refs. [15, 17]. In most of the cases the results of present work are in between the two earlier results. However, the difference between all three sets of results is small, $\sim 1\%$. This is consistent with the estimate of accuracy based on similar calculations for Pb (see previous section).

IV. CONCLUSION

We apply a recently developed advanced method of atomic structure calculation which combines three different all-order techniques to calculate energy levels of superheavy elements E113, Fl and Fl $^+$ with the accuracy $\sim 1\%$. This represents some improvement to previous calculations and contributes to the reliability of the

theoretical predictions of the spectra of superheavy elements.

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